Operational mapping of atmospheric nitrogen deposition to the Baltic Sea

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Operational mapping of atmospheric nitrogen deposition to the Baltic Sea

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Abstract

A new model system for mapping and forecasting nitrogen deposition to the Baltic Sea has been developed. The system is based on the Lagrangian variable scale transport-chemistry model ACDEP (Atmospheric Chemistry and Deposition model), and aims at delivering deposition estimates to be used as input to marine ecosystem models. The system is tested by comparison of model results to measurements from monitoring stations around the Baltic Sea. The comparison shows that observed annual mean ambient air concentrations and wet depositions are well reproduced by the model. Diurnal mean concentrations of NH_x (sum of NH_3 and NH_4^+) and NO_2 are fairly well reproduced, whereas concentrations of total nitrate (sum of HNO_3 and NO_3^-) are somewhat overestimated by the model. Wet depositions of nitrate and ammonia are fairly well described for annual mean values, whereas the discrepancy is high for the monthly mean values and the wet depositions are rather poorly described concerning the diurnal mean values. The model calculations show that the atmospheric nitrogen deposition has a pronounced south – north gradient with depositions in the range about 1.0 tonnes N km^{-2} in south and 0.2 tonnes N km^{-2} in north. The model results show that in 2000 the maximum deposition to the Danish waters appeared during the summer in the algae growth season. For the northern parts of the Baltic the highest depositions were distributed over most of the year.

1. Introduction

From the beginning of the 19th century and up to the middle of the 1980's, the nutrient input of nitrogen and phosphorous to the Baltic have increased by a factor of four and eight, respectively (Larsson et al., 1985). Oxygen deficits and subsequent death of fish and benthic fauna have become frequent phenomena over the same period of time (Meyer-Reil and Köster, 2000). These phenomena are directly linked to large algae production resulting from the high nutrient inputs (Rydberg et al., 1990), where

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nitrogen is considered to be the limiting factor in the coastal region (Paerl, 1995). When large amounts of dead algae deposit at the bottom, the oxygen in the bottom water of the sea is consumed in the degradation of the algae. Møhlenberg (1999) estimated that a 25% reduction in nitrogen to the Danish estuaries would lead to a 50% reduction in the number of days with severe oxygen depletion.

Besides playing a significant role in oxygen depletion episodes, it has been suggested in a number of papers that high nutrient inputs are responsible for an increased frequency of episodes with high concentrations of algae that are harmful to the health of humans and animals. However, the identification of harmful algae blooms is complex and there are no long time series of the occurrence in such episodes. A documentation of an increase in the occurrence and a link between this increase and high anthropogenic nutrient inputs has therefore not yet been given (Richardson, 1997).

Despite of its clear significance for the overall nitrogen loads to coastal waters like the Baltic, the atmospheric input has often been roughly determined and given little focus. Rosenberg et al. (1990) estimated that about 50% of the nitrogen load arise from atmospheric deposition. The main part of the atmospheric deposition is related to wash out of aerosol phase nitrogen compounds during rain events. Lindfors et al. (1993) found that dry deposition contributed to between 10 and 30% of the atmospheric nitrogen input to the Baltic. It has been suggested that events of high atmospheric nutrient inputs resulting from rain events may cause short- term blooms of algae under certain circumstances (Spokes et al., 1993, 2000). There is thus a need for high quality and high-resolution atmospheric nitrogen deposition estimates for use as input for marine ecosystem models.

In this paper a newly developed model system for producing high-resolution mapping as well as forecasts of nitrogen deposition to the Baltic Sea is presented. Data from this system will in turn be used as input for marine ecosystem models and the results obtained from the coupling will be published in subsequent papers.

2. The prognostic model system

Since summer 1998 the THOR forecasting system has been operated at the National Environmental Research Institute (NERI) (Brandt et al., 2000, 2001a, 2001b). The THOR system produces 3-days forecasts of air pollution on regional and local scale with focus on the Danish area. The Eta model (Nickovic et al., 1998) provides the meteorological forecasts that serve as input for the air pollution models. The air pollution models include the regional scale Danish Eulerian Operational Model (DEOM) (Brandt et al., 2001a; a validation is given in Tilmes et al., 2001), the urban scale Urban Background Model (UBM) (Berkowicz, 2000a) and the street scale Operational Street Pollution Model (OSPM) (Berkowicz et al., 1997; Berkowicz, 2000b).

Atmospheric nitrogen and sulphur depositions to Danish land and sea surfaces are calculated with the ACDEP (Atmospheric Chemistry and Deposition) model (Hertel al., 1995) on routine basis within the Danish National Background Monitoring Programme (DNBMP) (Ellermann et al., 2002). Meteorological parameters for the calculations are provided from the Eta model and initial concentrations from the DEOM, both operated under the THOR system. The calculations in DNBMP are performed for 233 receptor points in a 30 km×30 km grid and the results are carefully validated by comparison with measurements from the monitoring stations. In the present work the receptor net from DNBMP has been extended to cover the entire Baltic Sea area. This new receptor net contains in total 690 receptor points. Within the DNBMP the ACDEP model is operated in hind cast mode only, but in the present calculations for the Baltic Sea, the calculations are performed in both hind cast and forecast mode. The forecast computations are performed at 0500 each day, the results are stored and selected results are automatically uploaded to an FTP server available for the institutes that will run the marine ecosystem models.

ACDEP is a trajectory model where transport, chemical transformations and depositions are computed following an air parcel along 96 h back-trajectories. The air parcel is divided into 10 vertical layers from the ground and up to 2 km height. Transport of the entire air column is assumed to follow the σ-level 0.925 wind (approximately 800 m) disregarding wind turning with height.

The dry deposition velocity is described with the resistance method as given in Wesely and Hicks (1977). The aerodynamic resistance is computed with a standard method based on the relationship between wind speed, stability and the friction velocity. The laminar boundary layer resistance is given as a function of friction velocity, surface roughness and a surface roughness parameter of the specie. For land surfaces a constant surface roughness of 30 cm is assumed. For sea surfaces a slightly modified Charnock's formula is applied (Lindfors et al., 1991; Asman et al., 1994) so that the interdependence between friction velocity and sea surface roughness is taken into account. The roughness parameter for gaseous compounds is computed using formulas proposed by Brutsaert (1982), while for particles a method based on Slinn and Slinn (1980) is applied. The surface resistance over sea is modelled taking into account solubility and reactivity of the species in water (Asman et al., 1994).

The wet deposition is calculated taking into account both in-cloud and below cloud scavenging applying specific scavenging coefficients for the compounds in the model. It is assumed that in-cloud scavenging takes place in the model layers between 250 m and 2 km, while below cloud scavenging takes place in the layers below 250 m. The scavenging coefficients are computed taking into account solubility and wet phase reactivity (Hertel et al., 1995 and references herein). Depending on the rain intensity it is assumed that a larger or smaller fraction of a grid cell is covered by rain. This fraction is calculated applying a method described in Sandnes (1993).

The chemical module in the model is an extended version of the Carbon Bond Mechanism IV (CBM-IV) (Gery et al., 1989a, b) containing 35 chemical species and 80 chemical reactions. The extensions of the mechanism concern the description of ammonia and its reaction products. The numerical solver for the chemistry is the Eulerian Backward Iterative (EBI) method (Hertel et al., 1993), which has recently been considered to have the best accuracy/speed ratio among a variety of commonly applied solvers (Huang and Chang, 2001). However, the chemistry and the vertical diffusion

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are now solved simultaneously using the EBI method. This modification of the numerical treatment reduced the calculation time and improved the numerical accuracy considerably.

Accounting for horizontal dispersion in Lagrangian models requires analysis of many trajectories, which is highly computer demanding. A parameterisation has therefore been implemented to indirectly account for horizontal dispersion (Hertel et al. 1995). In average a plume grows by 1/10 of the travel distance from a source point. The emissions received by the air parcel are therefore averaged over an area around the centreline of the trajectory. This area has the width of 1/10 of the remaining distance along the trajectory to the receptor point.

3. Validation of the model

Model results have been compared to measurements from the EMEP monitoring stations around the Baltic Sea. The comparison is performed for 1999 since input data for the ACDEP calculations obtained from the THOR system is available only for 1999 and forward, and the latest available monitoring data from EMEP are from 1999. The aim of the performed comparison is to explore the ability of the model to reproduce annual, monthly and diurnal mean values. In the DNBMP the ACDEP model has only been compared to monitoring data on monthly and annual averages. However, the presented model system aim at producing data with a time resolution sufficient for describing nitrogen inputs on a time scale at which algal blooms take place. Such blooms may build up within a few days (Spokes et al., 1993).

3.1. Annual mean values

The comparison on annual mean basis is shown in Table 1. The comparisons show that the model tends to overestimate annual mean concentrations of nitrogen dioxide (about 10% in average) and total nitrogen (sum of HNO_3 and NO_3^-) (about 40% in average). Whereas NH_x (sum of NH_3 and NH_4^+) is underestimated (about 20% in average) (see also the graphical presentation in Fig. 1). The correlation between observed and computed concentrations is, however, generally high for all three species (0.78, 0.75 and 0.80, respectively). A good correlation indicates that the spatial distribution of the concentrations is described fairly well. Sulphate plays an important role in the atmospheric transport of ammonium. On average the model reproduces annual sulphate levels well, but the correlation between modelled and observed sulphate concentrations is relatively poor (0.47). Better correlation is obtained for sulphur dioxide (0.65), but here the concentrations are generally overestimated (see also Fig. 2).

The atmospheric nitrogen input to the North Sea is strongly dominated (about 80% on annual basis) by the contribution from wet deposition (Hertel et al., 2002). The model comparison shows that nitrate concentrations in precipitation (correlation of 0.90) are well reproduced (Table 1 and Fig. 3), although there is a tendency for a slight overestimation (in average about 20%). The correlation is smaller but still fair for ammonium in precipitation (0.65), but here with a similar tendency for underestimation (in average also about 20%). The same tendency is seen for the amount of wet deposition of the two compounds (Fig. 4).

3.2. Analysis of time series

Until now observed and computed annual mean values at the EMEP monitoring stations have been compared. In the following we will focus on time series and look into the model performance evaluated for monthly and diurnal mean values.

Figure 5 shows observed and modelled diurnal mean concentrations of NH_x, total nitrate and nitrogen dioxide averaged over all available monitoring stations. NH_x and nitrogen dioxide is generally well reproduced, whereas there is a general tendency to overestimate total nitrate. This result is in accordance with the comparisons performed on annual mean values for the single stations.

The analysis is expanded to investigate time series of correlation coefficients for the spatial distribution performed for the included 16 monitoring stations (see the description in Appendix 1). Figure 6 shows the correlation between observed and modelled diurnal mean values, monthly mean values and the annual mean values. For all three species high correlation is obtained for monthly (0.67 to 0.9, 0.56 to 0.9 and 0.75 to 0.9, respectively) and annual mean values (0.77, 0.75 and 0.79, respectively). Although fairly high correlations (above 0.5) are obtained for the main part of the time, weak correlations (below 0.4) are frequently obtained when diurnal mean values are evaluated.

The frequency of low nitrate and ammonium concentrations in precipitation (< 0.17 mg N/l) is higher in the model results than in observations (Fig. 7). For nitrate the total wet deposition is on average somewhat higher than observed due to a few modelled deposition events with high depositions (the plot is not shown here). The precipitation is in general well described by the model, although there are some of the observed episodes that are not reproduced.

On annual basis the modelled and observed ammonium and nitrate wet depositions are fairly well correlated (0.58 and 0.71). Already on monthly basis the picture is considerably more scattered and on diurnal basis the results are rather poor (Fig. 8). A significant part of the explanation may be found in the uncertainties in precipitation (also shown in Fig. 8). High correlations between observations and model results dominate when the annual mean values are evaluated, but even for monthly values a significant part of the results have correlations below 0.4.

3.3. Discussion

The model still resolves poorly wet depositions on short averaging times like diurnal means. It is likely that dry depositions are similarly uncertain, although a high correlation between observed and modelled ambient air concentrations is generally obtained. Several explanations may be given for this discrepancy of which the most important are believed to be uncertainties in:

– The emission data used for the model calculations,

- the precipitation data from the forecast model,
- the parameterisation of dry deposition processes,
- the parameterisation of aerosol processes, and
- general limitations associated with the principles of the Lagrangian model which we have applied.

Considering emission data, the uncertainties in annual emissions on 50 km×50 km EMEP grid have previously been estimated by EMEP to be in the order of 30 to 40%. However, even though the procedures around the submission of national emission data to the databases are well described, the are still data included in the databases that are subject to future corrections (Vesteng and Klein, 2002). These uncertainties increase further when data are distributed on sub-grid of 16.67 km×16.67 km and especially when highly simplified functions are applied for describing the seasonal and diurnal variation in emissions. We have initiated work that aim at improving the seasonal variation, especially concerning ammonia from agricultural activities.

The current application of the Eta model, which provides the meteorological input data for the model calculations, does not take into account detailed land use information. Land use has a significant impact on the distribution of precipitation. Figure 9 shows a comparison of gridded precipitation on 10 km×10 km provided by the Danish Meteorological Institute and similar figures obtained from the Eta model. The results show that the computed precipitation amounts are within the right order of magnitude, but the model results are more evenly distributed over the Danish land areas compared with the analysed precipitation data. The reason for this discrepancy is most probably that the surface topographic details are not sufficiently resolved in the relatively coarse resolution in the currently applied version of Eta. However, these issues are subjects of an ongoing project at NERI.

The current model does not take into account seasonal variation in land cover, and furthermore land use is only distributed on sea and land surfaces, where the latter

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is assumed covered by grass. A detailed land use database is in process of being implemented together with surface resistances for various land use types combined with information for the entire model domain (the EMEP area) about growing seasons, type of crops etc.

The model handles in general aerosol compounds in the same way as gaseous species. When dry deposition is considered, the aerosols are assumed to have a diameter of 0.8 μm. A new parameterisation is in process of being implemented. In this parameterisation aerosol size distributions are taken into account, and this is likely to improve the model performance although many parameters need to be determined in this context.

The Lagrangian model type has the advantage of being relatively little computer demanding, especially when a limited number of receptor points are considered. Furthermore the model scale may be changed along the trajectory, e.g. allowing for higher resolution in input data when the air parcel is approaching the receptor point. However, the uncertainty in the description of the transport may be significant in this type of models, especially considering the first part of the 96 h back-trajectory. Furthermore, wind turning with height is disregarded in the model, which may be a rather crude assumption. The next generation of nitrogen deposition model at NERI will therefore be a nested grid Eulerian model (Frohn et al., 2001; 2002).

4. Nitrogen depositions to the Baltic Sea

Episodes of high atmospheric nitrogen deposition are solely the result of precipitation events. Depositions may be somewhat elevated close to the coast when transport from nearby agricultural activities lead to high ammonia concentrations. However, the resulting dry deposition is considerably smaller than what is observed from rain out of aerosol phase ammonium and nitrate. Figure 10 shows the simulation of an event with high local wet deposition of atmospheric nitrogen in a belt from the coast of Poland and out to Gotland in the Baltic Sea. When the different plots are compared it is clear that the high deposition appears where there is an overlap between high aerosol phase concentrations and high precipitation amounts.

The computed total atmospheric nitrogen deposition to the Baltic Sea in 1999 is shown in Fig. 11. The deposition has a pronounced south – north gradient with depositions in the range about 1.0 tonnes N km^{-2} in south and 0.2 tonnes N km^{-2} in north. This gradient is due to transport from the areas with high emission density in the northern part of the European continent.

According to the model results, the maximum depositions over the Danish waters took place in the mid summer period where the algae growth is high (Fig. 12). For the northern part of the Baltic maximum values were distributed over most of the year. These results are again strongly dependent on the prediction of precipitation events and therefore rather uncertain.

5. Conclusions

The aim of the evaluation of the model performance was here to investigate how well the model reproduces air concentrations and wet depositions when short averaging times are considered. The results have shown that the model very well reproduces annual and monthly mean ambient air concentrations. Diurnal mean concentrations of NH_x (sum of NH_3 and NH_4^+) and NO_2 are fairly well reproduced, whereas total nitrate (sum of HNO_3 and NO_3^-) is somewhat overestimated by the model. Wet depositions of nitrate and ammonia are fairly well described for annual mean values, whereas the uncertainty is high for the monthly mean values and the wet deposition is poorly described for diurnal mean values.

The model calculations show that annual nitrogen depositions to the Baltic are in the range from 1 tonnes N km^{-2} in the south to 0.2 tonnes N km^{-2} in the north. Maximum diurnal depositions in 1999 seem to appear in the summer period for the Danish waters, but seem also to appear at any time of year for the rest of the Baltic. This result is quite uncertain and may only apply to this specific year.

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Appendix A

Monitoring stations used in the model validation

The following monitoring EMEP stations were included in the model validation for the year 1999 (further information may be obtained from http://www.nilu.no/projects/ccc/index.html). The station codes in Table A.1 is used in several of the plots and the situation of the stations is shown in Fig. A.1.

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Table 1. Comparison of observed and modelled ambient air concentrations (μg N/m^3 for the nitrogen compounds and μg S/m^3 for the sulphur compounds), concentrations in precipitation (μg/l) and precipitation (mm) at the 16 selected EMEP stations situated around the Baltic Sea. Comparisons of annual mean values for the years 1999. The number of data is indicated by n

Compound	Correlation	Maximum		Minimum		Mean		n
		Obs.	Mod.	Obs.	Mod.	Obs.	Mod.	
NO_2	0.78	2.12	2.74	0.62	0.87	1.42	1.58	10
$HNO_3+NO_3^-$	0.75	1.30	1.44	0.19	0.56	0.67	1.07	10
$NH_3+NH_4^+$	0.80	3.73	2.58	0.59	0.56	1.73	1.30	10
SO_2	0.65	1.46	2.46	0.37	0.67	0.72	1.33	12
SO_4^{2-}	0.47	1.25	1.45	0.31	0.56	0.82	0.87	11
Wet NH_4^+	0.65	0.63	0.60	0.18	0.13	0.44	0.37	12
Wet NO_3^-	0.90	0.61	0.83	0.26	0.26	0.47	0.56	12
Wet SO_4^{2-}	0.44	0.76	0.92	0.29	0.22	0.59	0.62	12
Precipitation	0.70	855	1072	339	530	640	719	12

Table A 1. Code of the monitoring stations used in the model validation. The code is given together with the name of the site, geographic coordinates and altitude about sea level. The situation of the stations is shown in Fig. A.1

Site	Code	Geographic coordinates	Altitude above sea level (m)
Country: Denmark			
Tange	DK03	56°21′ N, 9°36′ E	13
Keldsnor	DK05	54°44′ N, 10°44′ E	9
Anholt	DK08	56°43′ N, 11°31′ E	40
Country: Finland			
Ähtäri	FI04	62°33′ N, 24°13′ E	4
Virolahti II	FI17	60°31′ N, 27°41′ E	4
Country: Lithuania			
Preila	LT15 (SU15)	55°21′ N, 21°04′ E	5
Country: Latvia			
Rucava	LV10 (SU10)	56°13′ N, 21°13′ E	18
Country: Poland			
Leba	PL04	54°45′ N, 17°32′ E	2
Diabla Gora	PL05	54°09′ N, 22°04′ E	157

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Table A 1. Continued

Site	Code	Geographic coordinates	Altitude above sea level (m)
Country: Sweden			
Rörvik	SE02	57°25′ N, 11°56′ E	10
Hoburg	SE08	56°55′ N, 18°09′ E	58
Vavihill	SE11	56°01′ N, 13°09′ E	172
Aspvreten	SE12	58°48′ N, 17°23′ E	20
Country: Estonia			
Lahemaa	EE09 (SU09)	59°30′ N, 25°54′ E	32
Vilsandi	EE11 (SU11)	58°23′ N, 21°49′ E	6
Country: Germany			
Zingst	DE09	54°26′ N, 12°44′ E	1

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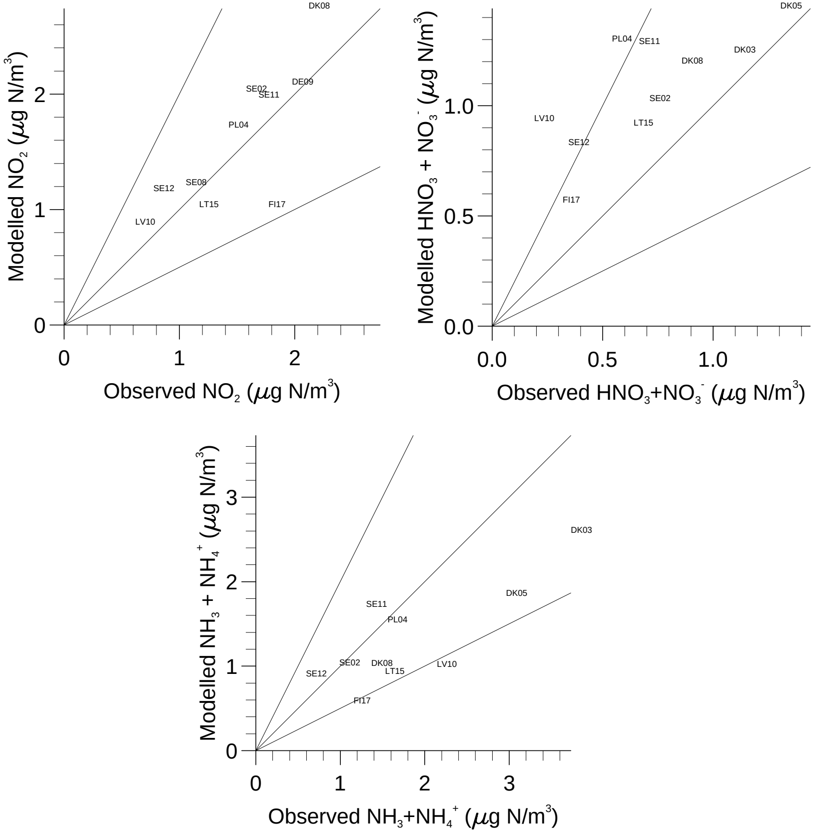

Fig. 1. Comparison between observed and calculated nitrogen dioxide, the sum of nitric acid and aerosol phase nitrate, and NH_x (the sum of NH_3 and NH_4^+) at the 16 selected stations in the EMEP programme. Annual mean values for 1999. The stations in the figure is explained in Appendix 1 and strait lines indicate 1:1, 1:2 and 2:1, respectively.

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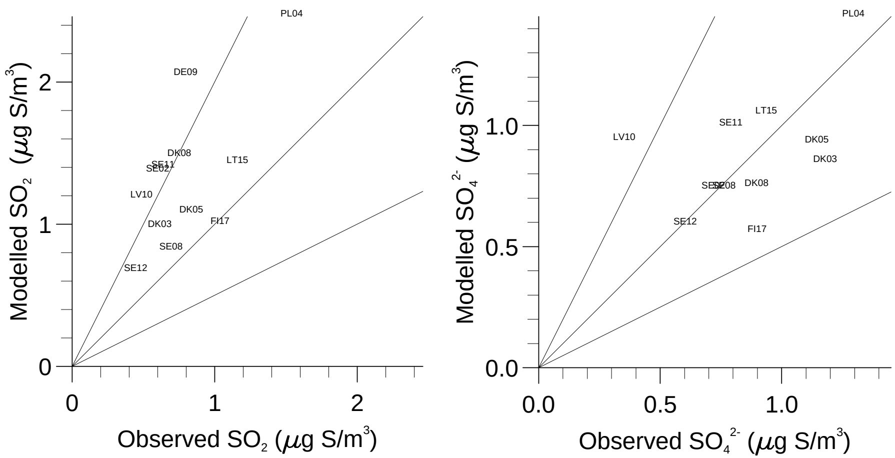

Fig. 2. Comparison between observed and calculated sulphur dioxide and sulphate at the 16 selected stations in the EMEP programme. Annual mean values for 1999. The stations in the figure is explained in Appendix 1 and strait lines indicate 1:1, 1:2 and 2:1, respectively.

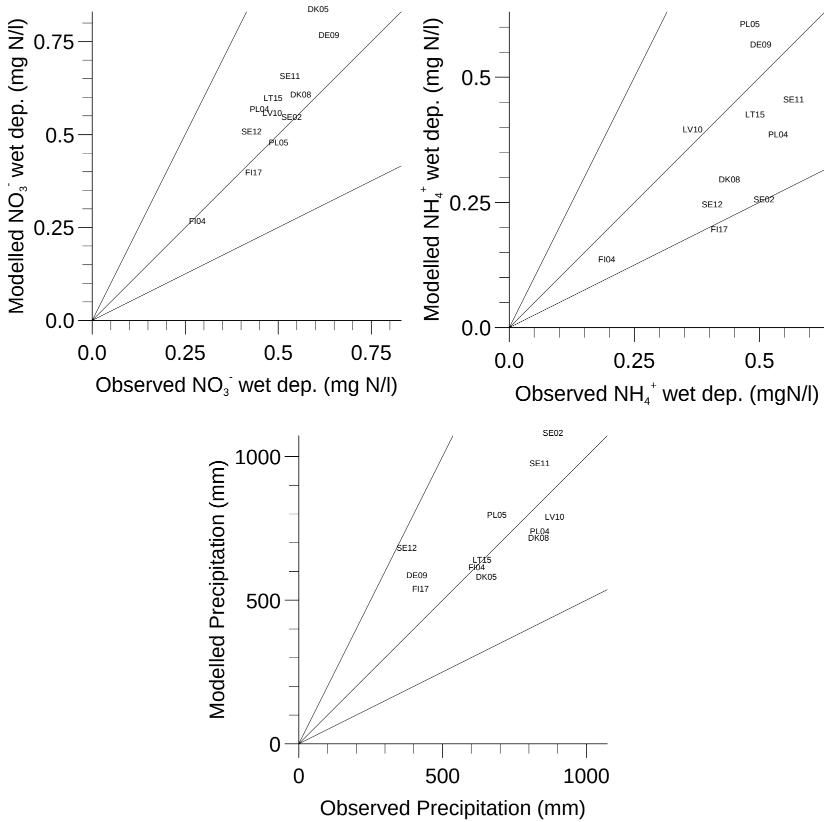

Fig. 3. Comparison between observed and calculated concentrations in precipitation of nitrate and ammonium, and of observed and calculated precipitation at the 16 selected stations in the EMEP programme. Annual mean values for 1999. The stations in the figure is explained in Appendix 1 and strait lines indicate 1:1, 1:2 and 2:1, respectively.

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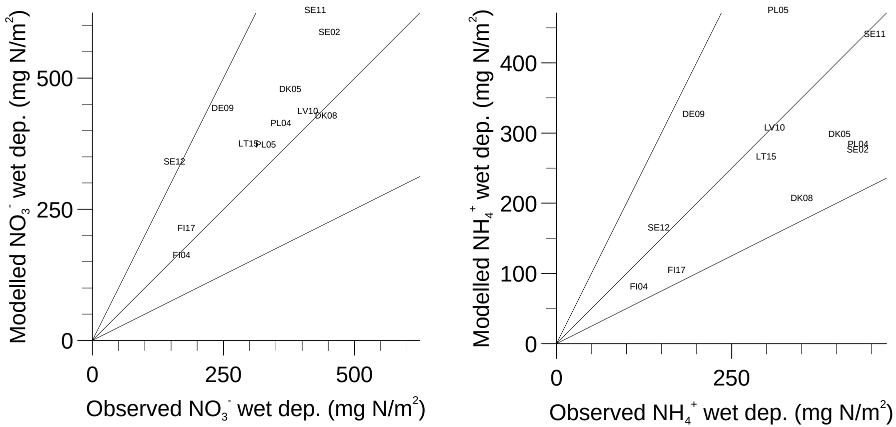

Fig. 4. Comparison between observed and calculated wet deposition of nitrate and ammonium at the 16 selected stations in the EMEP programme. Annual mean values for 1999. The stations in the figure is explained in Appendix 1 and strait lines indicate 1:1, 1:2 and 2:1, respectively.

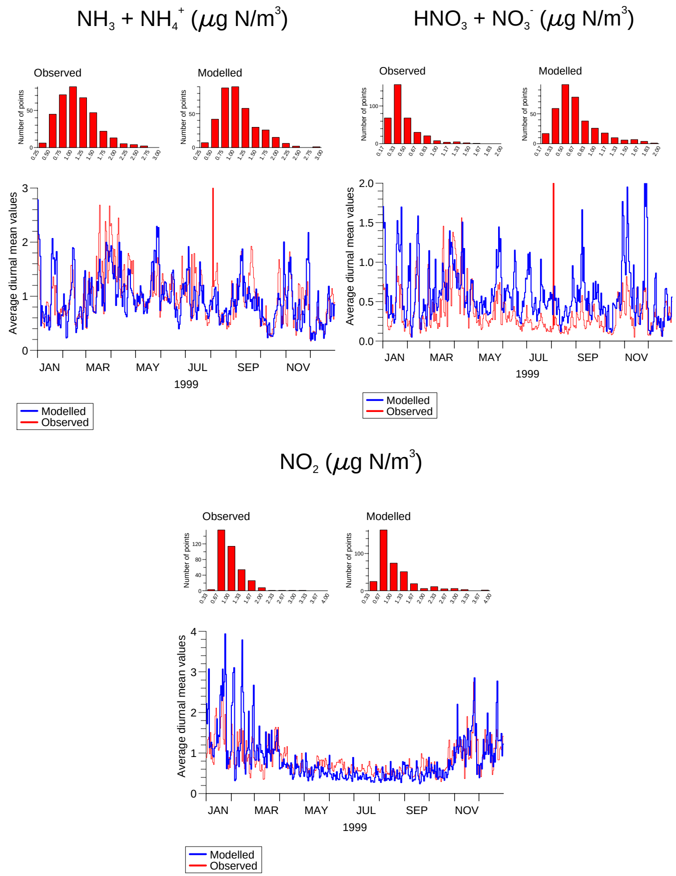

Fig. 5. Comparison of average concentrations of NH_x (sum of NH_3 and NH_4^+), total nitrate (sum of HNO_3 and NO_3^-) and NO_2 for diurnal mean values – all data for the year 1999. The averaging is performed over the 16 selected EMEP stations for each set of data.

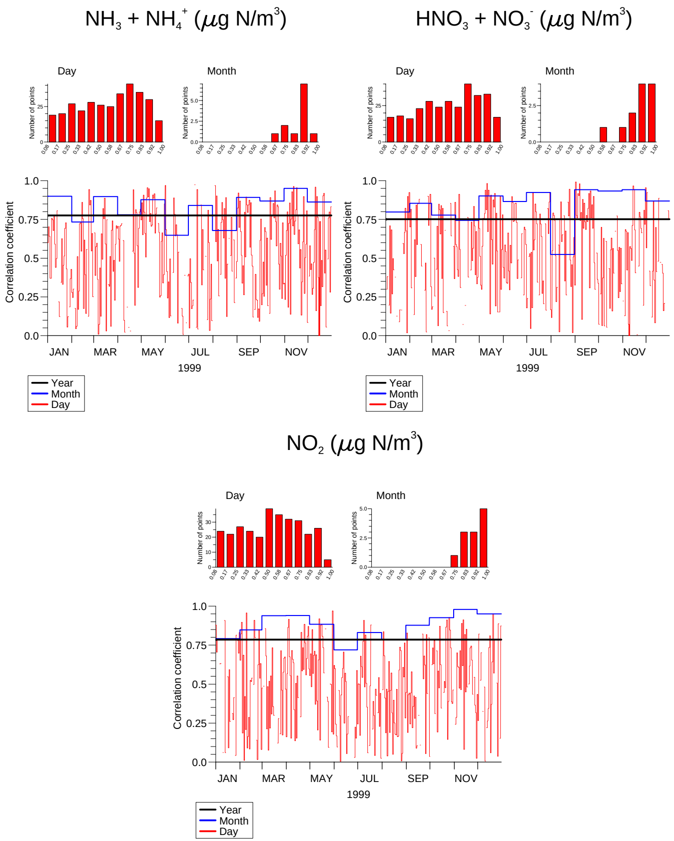

Fig. 6. Correlation coefficients between observed and modelled NH_x (sum of NH_3 and NH_4^+), total nitrate (sum of HNO_3 and NO_3^-) and NO_2 for annual, monthly and diurnal mean values – all data for the year 1999. The averaging is performed over the 16 selected EMEP stations for each set of data.

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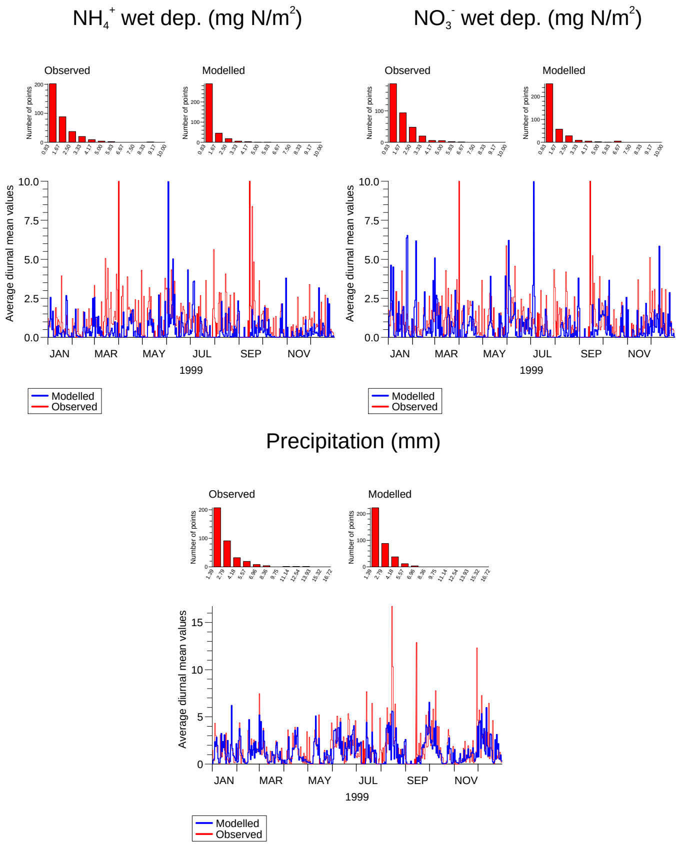

Fig. 7. Comparison of average wet depositions of NH_4^+ and NO_3^-, and precipitation for diurnal mean values in 1999. The averaging is performed over the 16 selected EMEP stations for each set of data.

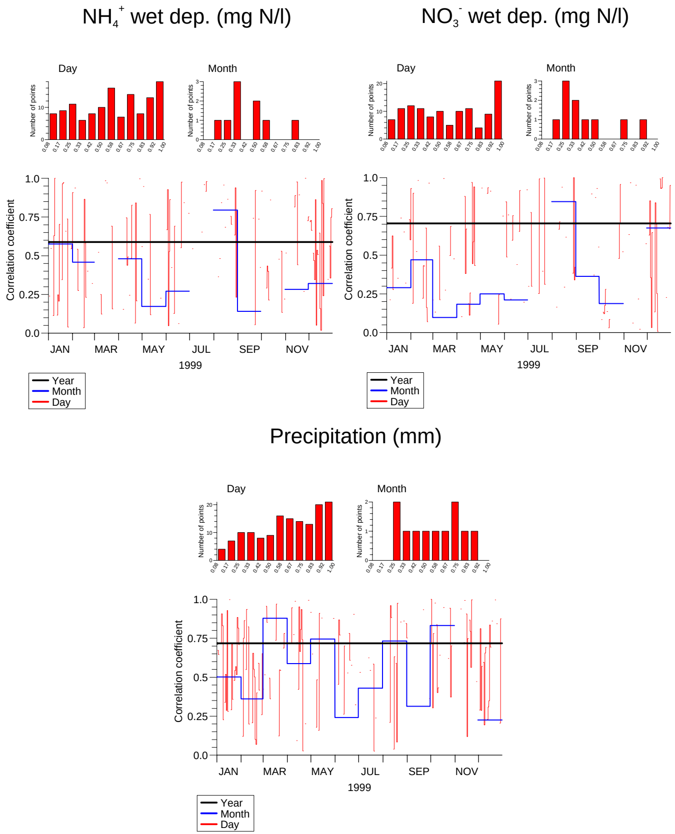

Fig. 8. Correlation coefficients for concentrations in precipitation of NH_4^+ and NO_3^-, and precipitation for diurnal mean values for the year 1999. The averaging is performed over the 16 selected EMEP stations for each set of data.

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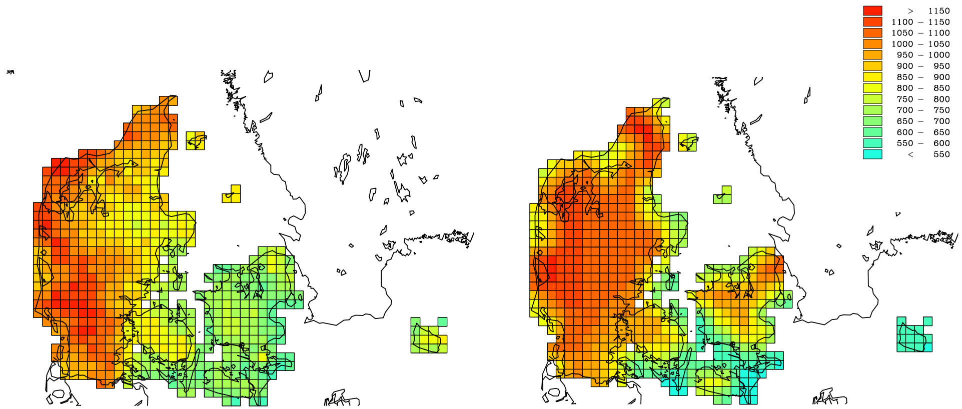

Fig. 9. Gridded precipitation on 10 km×10 km gridded from observed precipitation data (left) and obtained from the Eta model (right). Measurements have been provided by the Danish Meteorological Institute (Scharling, 1998).

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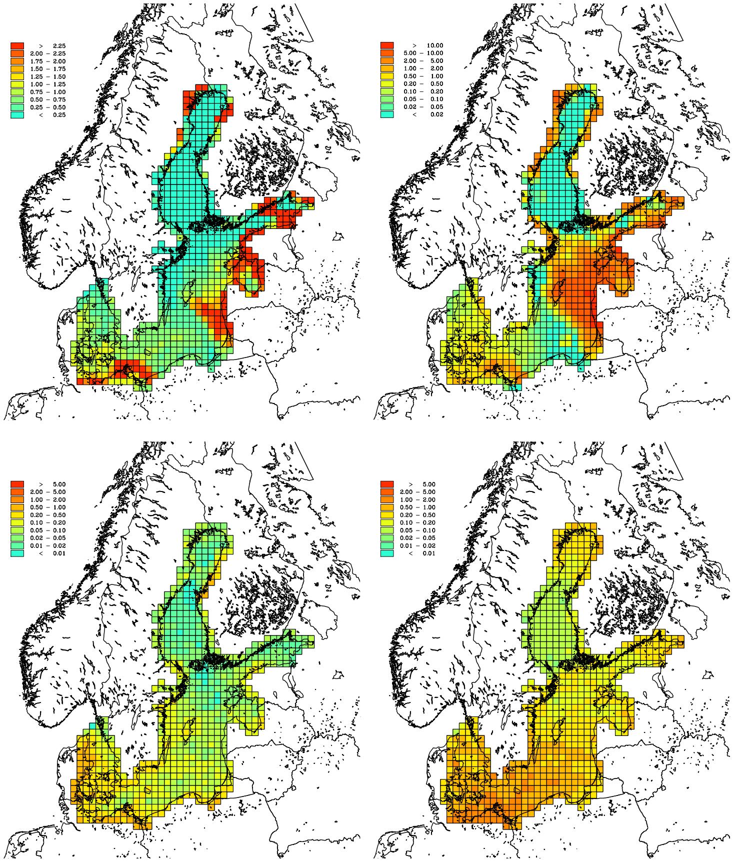

Fig. 10. Episode of high nitrogen deposition at the 26th of July 2002. The upper left figure shows the total nitrogen deposition (tonnes N/km^2) on this day. The upper right figure shows the precipitation (mm). The lower left figure shows the concentration of ammonia (μg N/m^3) and the lower right figure the concentration of particulate ammonium (μg N/m^3).

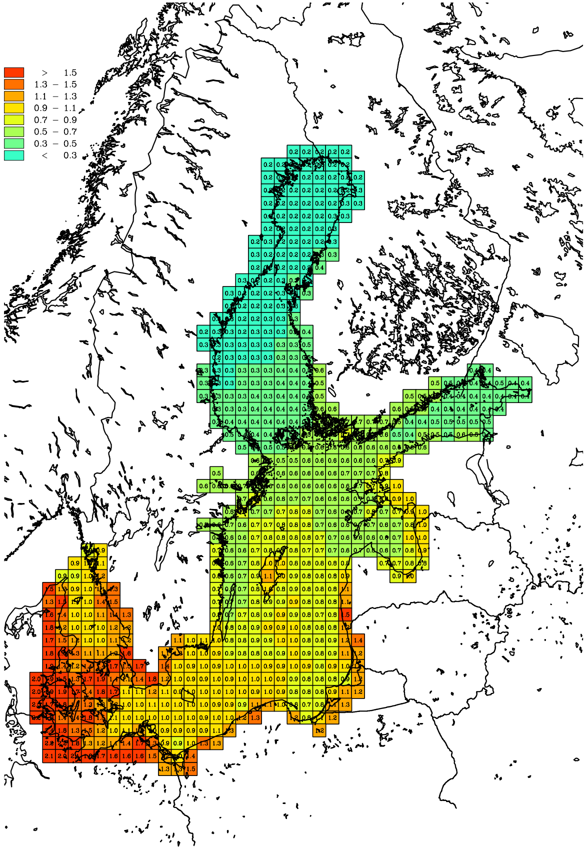

Fig. 11. Calculated nitrogen deposition (tonnes N km^{-2}) to the entire Baltic Sea in 1999.

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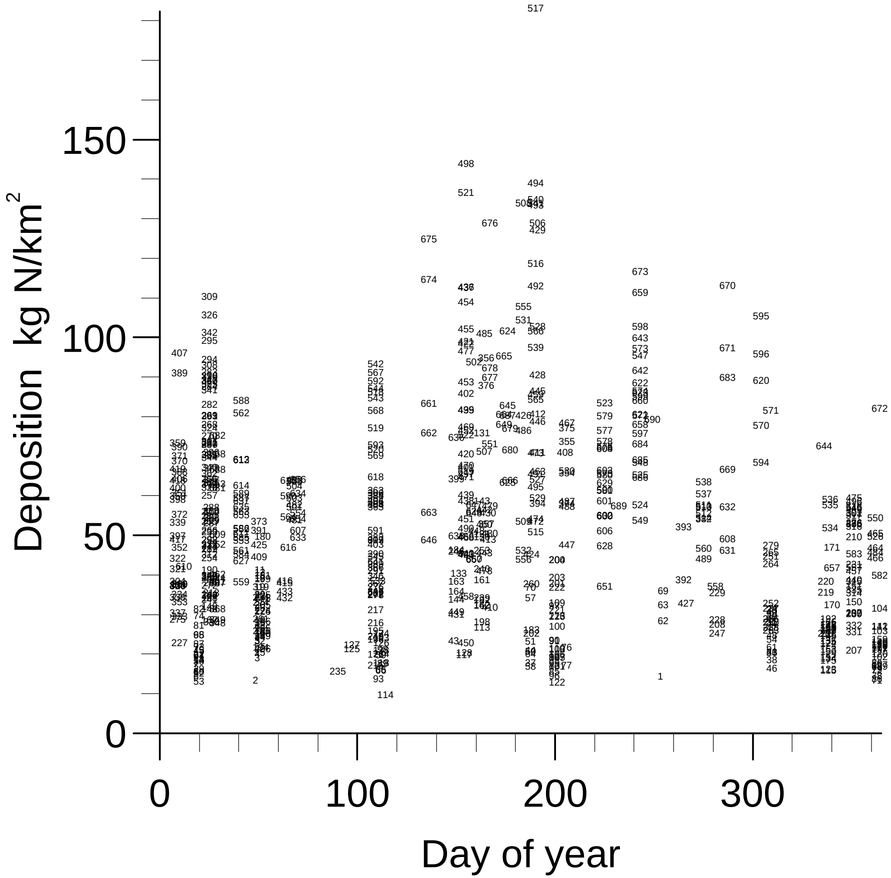

Fig. 12. Calculated maximum diurnal nitrogen deposition density (kg N km^{-2}) to 690 receptor points distributed over the entire Baltic Sea in 1999. The numbers in the plot refer to the receptor grid number, which starts in the most northern part of the Baltic and ends with the Danish waters.

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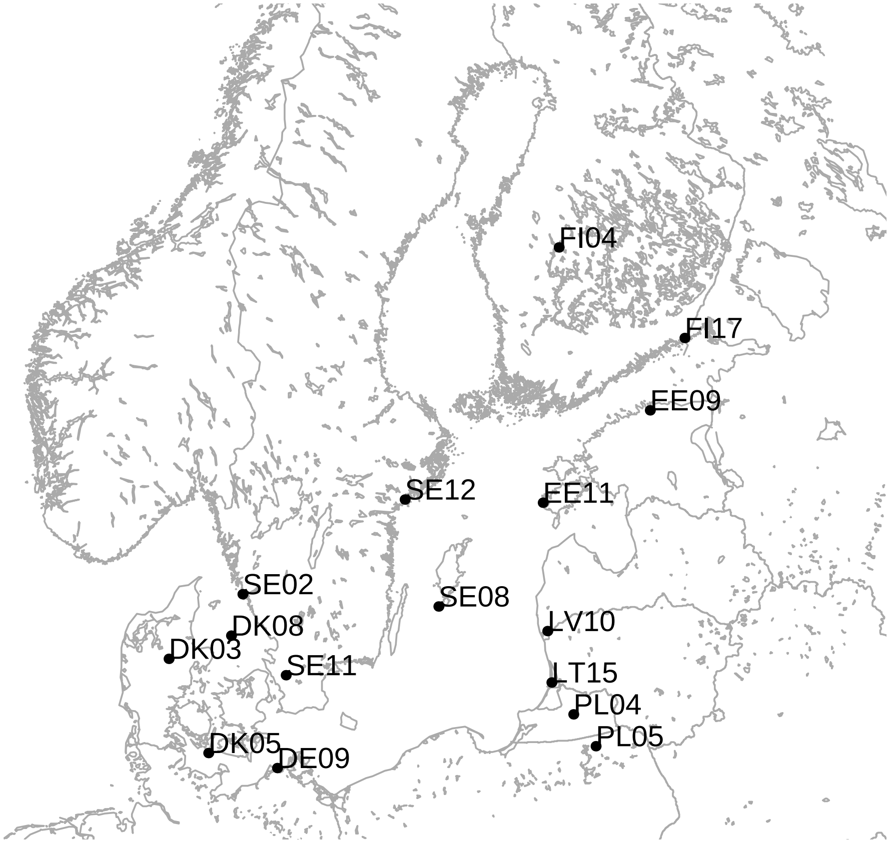

Figure A 1. Situation of the monitoring stations used in the model validation. The names of the sites, the geographic coordinates and the altitude above sea level are given in Table A.1.

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